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(54) HONEYCOMB CERAMICS FILTER

(57) There is disclosed a honeycomb ceramics filter, wherein an average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy a relation of $10 \geq W/X$. According to this filter, while a predetermined

trapping efficiency is maintained, a pressure loss can be prevented from increasing to be not less than a predetermined pressure loss even with use for a long period.

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Description**TECHNICAL FIELD**

[0001] The present invention relates to a honeycomb ceramics filter which has a honeycomb shape. The present invention further particularly relates to a honeycomb ceramics filter in which trapping efficiency of particulates is sacrificed to some extent, but an ash content generated by oil combustion of a diesel engine is extracted from pores so as to prevent a pressure loss from increasing to be not less than a predetermined loss even with a long-period use.

BACKGROUND ART

[0002] In recent years, a porous honeycomb ceramics filter has been used as an apparatus for removing particulates in exhaust gas, including a structure in which a plurality of through holes opened in end surfaces on exhaust gas inflow and outflow sides are alternately closed in the opposite end surfaces. The exhaust gas which has flown in from the end surface on the exhaust gas inflow side is forcibly passed through partition walls (including a plurality of fine pores) among the respective through holes to trap/remove the particulates in the exhaust gas.

[0003] For the ceramics filter, a pore diameter of the fine pore formed in the partition wall between the respective through holes is determined by a relation with a particulate diameter of the particulate in the exhaust gas, and capabilities such as trapping efficiency and pressure loss differ with a degree of the pore diameter. In view of this, development has heretofore been advanced so as to obtain the ceramics filter having a high trapping efficiency of particulates (such as soot).

[0004] However, for the filter having the raised trapping efficiency, since the pore diameter of the filter is small, there is a problem that an ash content generated by oil combustion of a diesel engine is accumulated in the pores of the filter and the pressure loss is raised to be not less than a predetermined loss by the use for a long period. For the filter having a small pore diameter, the pressure loss is high in sampling the particulates (such as the soot), and there has been a demand for reduction of the loss.

[0005] Additionally, in recent years, by improvement of the diesel engine, the particulates such as the soot generated from the engine have been reduced by a considerable amount than before. Then, as a result of the present inventor's intensive studies, in order to clear a regulated value of a future exhaust gas standard, it is not necessary to achieve an extremely high trapping efficiency. Conversely, it has been found that in order to prevent a rise of the pressure loss caused by the accumulation of the ash content into the pores, it is important to increase the pore diameter to such an extent that the exhaust gas can pass through the partition wall, and the present invention has been attained.

[0006] Therefore, an object of the present invention is to provide a honeycomb ceramics filter in which a predetermined trapping efficiency is maintained, an ash content generated by oil combustion of a diesel engine is extracted from pores, and pressure loss is accordingly prevented from increasing to be not less than a predetermined loss even with long-period use.

DISCLOSURE OF THE INVENTION

[0007] That is, according to the present invention, there is provided a honeycomb ceramics filter, characterized in that an average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

$$10 \geq W/X.$$

[0008] In the ceramics filter of the present invention, the average pore diameter X (μm) and partition wall thickness W (μm) of the filter preferably satisfy a relation of $7 \geq W/X \geq 3$. The average pore diameter X (μm) and partition wall thickness W (μm) of the filter more preferably satisfy a relation of $5 \geq W/X \geq 3$.

[0009] Moreover, in the ceramics filter of the present invention, a porosity is preferably 55 to 75%, and a ceramic preferably contains cordierite and/or silicon carbide as a major component. Furthermore, in the ceramics filter of the present invention, a thermal expansion coefficient at 40 to 800°C is preferably $1.0 \times 10^{-6}/^\circ\text{C}$ or less. The filter of the present invention preferably has a honeycomb shape whose partition wall thickness W is 350 μm or less and whose cell density is 250 cells/in² or more.

BEST MODE FOR CARRYING OUT THE INVENTION

[0010] Embodiments of the present invention will concretely be described hereinafter, but the present invention is not limited to these embodiments.

[0011] In the present invention, a pore diameter and partition wall thickness are controlled so that an average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy a relation of $10 \geq W/X$ to constitute a honeycomb ceramics filter.

[0012] As described above, the present inventor has found that it is important to increase a pore diameter to such an extent that exhaust gas can pass through partition walls in order to prevent a rise of pressure loss caused by accumulation of an ash content into pores. Concretely, the present inventor has intensively studied a relation between the pore diameter and the partition wall thickness. When the pore diameter is increased to be not less than a predetermined diameter with respect to the partition wall thickness of a honeycomb structure, trapping efficiency of particulates such as soot drops to some extent, but the pressure loss at the time of sampling the particulates can be reduced. Additionally, the ash content accumulated in the pores formed inside partition walls can be extracted from the pores. As a result, it has been found that it is possible to obtain a honeycomb ceramics filter which can stably function for a long period without increasing the pressure loss to be not less than the predetermined loss even with the long-period use.

[0013] In the present invention, when a relation W/X between the average pore diameter X (μm) and partition wall thickness W (μm) of the filter is larger than 10, the exhaust gas does not easily pass through the partition walls, and the ash content accumulated in the pores cannot be extracted from the pores.

[0014] In the present invention, the relation between the average pore diameter X (μm) and the partition wall thickness W (μm) is preferably $7 \geq W/X \geq 3$ in order to achieve the above-described pores, and more preferably $5 \geq W/X \geq 3$.

[0015] Moreover, in the honeycomb ceramics filter of the present invention, in order to achieve the above-described object of the present invention, the average pore diameter X is usually 20 to 70 μm , more preferably 30 to 70 μm . The partition wall thickness W is preferably 350 μm or less, and more preferably in a range of 200 to 300 μm .

[0016] Major components of the ceramics filter of the present invention are not especially limited, and any type of ceramic can be used, but cordierite and/or silicon carbide are preferably the major components. Cordierite may be oriented, non-oriented, α -crystalline, or β -crystalline. Silicon carbide may be either α -crystalline or β -crystalline.

[0017] Moreover, other components may also be contained such as mullite, zirconium, aluminum titanate, clay bond silicon carbide, zirconia, spinel, indialite, sapphirine, corundum, and titania.

[0018] The honeycomb filter of the present invention has a porosity of preferably 55 to 75%, more preferably 60 to 70% in terms of the reduction of the pressure loss and the trapping efficiency. In terms of enhancement of thermal shock resistance at the time of the use at a high temperature, a thermal expansion coefficient at 40 to 800°C is preferably $1.0 \times 10^{-6}/^\circ\text{C}$ or less, more preferably $0.8 \times 10^{-6}/^\circ\text{C}$ or less.

[0019] Moreover, the ceramics filter of the present invention is usually of a honeycomb type including a structure in which a plurality of through holes opened in end surfaces on exhaust gas inflow and outflow sides are alternately closed in the opposite end surfaces. However, the shape of the honeycomb filter is not especially limited, and any of shapes may be used such as a column whose end surface has a perfect circular or elliptic shape, a prism whose end surface has a triangular, quadrangular, or otherwise polygonal shape, and a column or prism whose side surface has a curved V-shape. Moreover, the shape of the through hole is not especially limited, and the section may have any of the shapes such as the polygonal shapes including the quadrangular and octagonal shapes, the perfect circular shape, and the elliptic shape. The filter has a cell density in a range of preferably 250 cells/in² or more, more preferably 300 to 400 cells/in² in terms of the sampling capability of the exhaust gas.

[0020] The honeycomb ceramics filter of the present invention can be manufactured by the following methods.

[0021] First, when a cordierite material is used as a starting material of the filter, and when the cordierite material is blended with components so as to obtain a theoretical composition of a cordierite crystal, it is necessary to blend silica (SiO_2) source components, magnesia (MgO) source components such as kaolin and talc, alumina (Al_2O_3) source components such as aluminum oxide and aluminum hydroxide, and the like.

[0022] For the alumina (Al_2O_3) source components, the material containing either one or both of aluminum oxide and aluminum hydroxide is preferable because of a small amount of impurities. Above all, it is preferable to contain aluminum hydroxide.

[0023] Moreover, for the alumina (Al_2O_3) source components, the cordierite material preferably contains 15 to 45% by mass of aluminum hydroxide, or 0 to 20% by mass of aluminum oxide.

[0024] Examples of the magnesia (MgO) source components include talc and magnesite. Above all, talc is preferable. The cordierite material preferably contains 37 to 40% by mass of talc, and a particle diameter of talc is preferably 5 to 40 μm , more preferably 10 to 30 μm in order to reduce the thermal expansion coefficient.

[0025] Moreover, the magnesia (MgO) source components such as talc for use in the present invention may contain Fe_2O_3 , CaO , Na_2O , K_2O , and the like which are impurities. Additionally, the content of Fe_2O_3 is preferably 0.1 to 2.5% by mass in the magnesia (MgO) source component. With the content in this range, the thermal expansion coefficient can be reduced, and a high porosity can also be obtained.

[0026] Furthermore, the contents of CaO , Na_2O , K_2O are preferably set to 0.35% by mass or less in total in the magnesia (MgO) source component so as to lower the thermal expansion coefficient.

[0027] It is to be noted that in the present invention, silicon carbide can be the major component as the starting material of the filter. The case in which silicon carbide is the major component includes both the cases in which silicon carbide (SiC) is the major component and in which silicon carbide (SiC) and metal silicon (Si) are the major components.

5 [0028] When the filter of the present invention is manufactured, the start material containing the cordierite material and/or silicon carbide as the major component may be blended with various additives if necessary. Examples of the additives include a foamed resin, a binder, a dispersant for promoting dispersion into a medium solution, and a hole making material for forming pores.

10 [0029] Examples of the foamed resin include acrylic microcapsule, and the like. Examples of the binder include hydroxypropyl methyl cellulose, methyl cellulose, hydroxyl ethyl cellulose, carboxyl methyl cellulose, polyvinyl alcohol, and the like. Examples of the dispersant include ethylene glycol, dextrin, fatty acid soap, polyalcohol, and the like. Examples of the hole making agent include graphite, flour, starch, phenol resin, polymethyl methacrylate, polyethylene, polyethylene terephthalate, and the like.

15 [0030] These additives can be used alone or as a combination of two or more thereof in accordance with purposes.

15 [0031] In the present invention, the above-described starting materials can be used to manufacture the honeycomb ceramics filter in the following manufacturing steps.

20 [0032] First, 3 to 5 parts by weight of the binder, 3 to 40 parts by weight of the hole making agent, 0.5 to 2 parts by weight of the dispersant, and 10 to 40 parts by weight of water are cast into 100 parts by weight of the above-described starting material, kneaded, and plasticized.

25 [0033] Subsequently, the plastic material can be molded by an extrusion molding method, an injection molding method, a press molding method, a method of molding a ceramic material into a columnar shape and thereafter forming through holes, and the like. Above all, the extrusion molding method is preferably carried out in that continuous molding is easy and, for example, a cordierite crystal can be oriented to obtain a low thermal expansion property.

25 [0034] Next, the molded material can be dried by hot-air drying, microwave drying, dielectric drying, reduced-pressure drying, vacuum drying, freeze drying, and the like. Above all, a drying step in which the hot-air drying is combined with the microwave drying or the dielectric drying is preferably carried out in that the whole material can quickly and uniformly be dried.

30 [0035] Finally, to calcine the dried/molded material, depending on the size of the dried/molded material, usually with the cordierite material, the material is preferably calcined under the atmosphere at temperature of 1410 to 1440°C for three to seven hours. With the material containing silicon carbide which is the major component, the material is calcined under a non-oxidizing atmosphere such as N₂ and Ar in order to prevent oxidation of SiC. When SiC is bonded by silicon nitride, the calcining temperature is a temperature at which a silicon nitride powder is softened, and the calcining is preferably carried out at a temperature of 1550 to 2000°C. When SiC particles are bonded to one another by a recrystallization method, it is necessary to calcine the material at a temperature of at least 1800°C or more. Furthermore, when SiC and Si are major components, the calcining is preferably carried out under the non-oxidizing atmosphere such as N₂ and Ar at a temperature of 1400 to 1800°C. It is to be noted that the drying and calcining steps may also continuously be carried out.

35 [0036] The present invention will more concretely be described by means of examples hereinafter, but the present invention is not limited to these examples.

40 1. Evaluation Method

[0037] The honeycomb ceramics filters obtained in Examples 1 to 13 and Comparative Examples 1 to 5 described later were evaluated by the following methods.

45 (1) Average Pore Diameter

[0038] The average pore diameter was measured by a mercury press-in porosimeter manufactured by Micromeritics Co.

50 (2) Porosity

[0039] A true specific gravity of cordierite was set to 2.52 g/cm³, and the porosity was calculated from a total pore volume. The true specific gravity of SiC was set to 3.05 g/cm³.

55 (3) Trapping efficiency

[0040] Exhaust gas from which soot was generated by a soot generator was allowed to flow into a filter having a size: φ144 mm × 152 mm (length) and having an average pore diameter X, partition wall thickness W, porosity, cell

density, and thermal expansion coefficient shown in Table 2 for two minutes. The soot included in the exhaust gas passed through the filter was trapd with filter paper, and a weight (W_1) of the soot was measured. At the same time, the exhaust gas from which the soot was generated was trapd with the filter paper without being passed through the filter, and a weight (W_2) of the soot was measured. Subsequently, the obtained weights (W_1) and (W_2) were assigned to the following equation to obtain the trapping efficiency:

$$(W_2 - W_1) / (W_2) \times 100.$$

10 (4) Soot Sampling Pressure Loss Evaluation

[0041] The honeycomb ceramics filter having the size: $\phi 144 \text{ mm} \times 152 \text{ mm}$ (length) was used, front and back of the honeycomb filter were pressed with a ring having an inner diameter of $\phi 130 \text{ mm}$, and measurement was carried out substantially in the inner diameter of $\phi 130 \text{ mm}$. The soot was generated by the soot generator, and 10 g of soot was trapd by the honeycomb filter. In this state, $2.27 \text{ Nm}^3/\text{min}$ of air was passed, and a pressure difference before/after the filter was measured.

(Examples 1 to 12)

20 [0042] Main materials and hole making materials were mixed to prepare various cordierite materials with average particle diameters and blend ratios shown in Table 1.

[0043] Subsequently, with respect to 100 g of each of these various cordierite materials, 4 g of hydroxypropyl methyl cellulose, 0.5 g of lauric acid potash soap, and 30 g of water were cast, kneaded, and plasticized. This plastic material was formed into a cylindrical mound of earth with a vacuum earth kneader, cast into an extrusion molding machine, and molded in a honeycomb shape.

[0044] Subsequently, various obtained molded materials were dielectrically dried, and thereafter absolutely dried by the hot-air drying, and opposite end surfaces were cut into predetermined dimensions.

[0045] Subsequently, the through holes in the honeycomb-shaped dried material were alternately closed in the opposite end surfaces in which the through holes were opened with a flurry formed of the cordierite material having the similar composition.

[0046] Finally, after the calcining at 1420°C for four hours, according to Examples 1 to 12 and Comparative Examples 1 to 5, the honeycomb ceramics filter having the size: $\phi 144 \text{ mm} \times 152 \text{ mm}$ (length) was obtained.

(Example 13)

35 [0047] The main materials (SiC and Si) and hole making materials shown in Table 1 were used to carry out the drying and hole closing in the same method as that of Examples 1 to 12. The materials were calcined below 400°C in an ambient-pressure oxidizing atmosphere, at 400°C or more in an ambient-pressure argon atmosphere, and at a maximum temperature of 1450°C for one hour to obtain the honeycomb ceramics filter having the size: $\phi 144 \text{ mm} \times 152 \text{ mm}$ (length).

40 [0048] Evaluation results of Examples 1 to 13 and Comparative Examples 1 to 5 are shown altogether in Table 2.

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Table 1

No.	Main material						Hole making material	
	Talc (% by mass)	Kaolin (% by mass)	Quartz (% by mass)	Molten silica (% by mass)	Aluminum oxide (% by mass)	Aluminum hydroxide (% by mass)	Graphite (% by mass)	Foamed resin (% by mass)
Example 1	40 (15 µm)	19 (10 µm)	12 (39 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	2 (52 µm)
Example 2	40 (15 µm)	19 (10 µm)	12 (39 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	2 (52 µm)
Example 3	40 (15 µm)	19 (10 µm)	12 (39 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	2 (52 µm)
Example 4	40 (15 µm)	0	21 (10 µm)	0	16 (7 µm)	23 (3 µm)	20 (29 µm)	4 (35 µm)
Example 5	40 (15 µm)	0	21 (10 µm)	0	16 (7 µm)	23 (3 µm)	20 (29 µm)	4 (35 µm)
Example 6	40 (15 µm)	0	21 (10 µm)	0	16 (7 µm)	23 (3 µm)	20 (29 µm)	4 (35 µm)
Example 7	40 (15 µm)	0	21 (10 µm)	0	16 (7 µm)	23 (3 µm)	20 (29 µm)	4 (35 µm)
Example 8	40 (7 µm)	0	0	21 (78 µm)	16 (5 µm)	23 (1.5 µm)	20 (29 µm)	4 (64 µm)
Example 9	40 (20 µm)	0	21 (89 µm)	0	16 (10 µm)	23 (1.5 µm)	0	3 (64 µm)
Example 10	40 (20 µm)	0	21 (89 µm)	0	16 (10 µm)	23 (1.5 µm)	0	3 (64 µm)
Example 11	40 (20 µm)	0	0	21 (54 µm)	16 (10 µm)	23 (2 µm)	0	2 (52 µm)
Example 12	40 (15 µm)	19 (10 µm)	0	12 (32 µm)	14 (7 µm)	15 (2 µm)	20 (29 µm)	4 (35 µm)
Example 13	SiC (35 µm): 80 % by mass, Si (6 µm): 20 % by mass							
Comparative Example 1	40 (15 µm)	0	21 (10 µm)	0	16 (7 µm)	23 (3 µm)	20 (29 µm)	4 (35 µm)
Comparative Example 2	40 (15 µm)	19 (10 µm)	12 (10 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	3 (52 µm)
Comparative Example 3	40 (15 µm)	19 (10 µm)	12 (10 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	3 (52 µm)
Comparative Example 4	40 (15 µm)	19 (10 µm)	12 (10 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	3 (52 µm)
Comparative Example 5	40 (15 µm)	19 (10 µm)	12 (10 µm)	0	14 (7 µm)	15 (2 µm)	15 (51 µm)	3 (52 µm)

Numerals within parentheses in the table denote the average particle diameters of the materials.

Table 2

No.	Partition wall thickness W (μm)	Cell density (/in ²)	Porosity (%)	Average pore diameter X (μm)	W/X	Thermal expansion coefficient ($\times 10^{-6}/^\circ\text{C}$)	Soot sampling pressure loss (KPa)	Trapping efficiency (%)
Example 1	300	300	59	32	9.4	0.6	9.4	86
Example 2	250	300	59	32	7.8	0.6	8.6	71
Example 3	200	300	59	32	6.3	0.6	7.3	59
Example 4	200	350	70	24	8.3	0.7	5.2	79
Example 5	150	350	70	24	6.3	0.7	4.9	58
Example 6	150	400	70	24	6.3	0.7	4.2	52
Example 7	100	400	70	24	4.2	0.7	3.5	45
Example 8	300	300	75	45	6.7	1.0	7.6	63
Example 9	300	300	61	61	4.9	1.0	9.3	48
Example 10	350	300	61	61	5.7	1.0	10.3	50
Example 11	200	300	55	45	4.4	0.5	7.9	45
Example 12	100	400	68	32	3.1	0.8	3.7	42
Example 13	300	250	58	31	9.7	4.1	7.3	89
Comparative Example 1	250	300	70	24	10.4	0.7	7.1	91
Comparative Example 2	200	300	68	15	13.3	0.8	6.8	94
Comparative Example 3	300	300	68	15	20.0	0.8	8.6	96
Comparative Example 4	400	300	68	15	26.7	0.6	10.3	97
Comparative Example 5	300	200	68	15	20.0	0.6	10.7	95

INDUSTRIAL APPLICABILITY

[0049] As described above, since a value obtained by dividing a partition wall thickness by an average pore diameter has correlation with a trapping efficiency, according to the present invention, the value obtained by dividing the partition wall thickness by the average pore diameter can be controlled to freely control the trapping efficiency. Since the trapping efficiency of soot is assumed to have correlation with that of an ash content, according to the present invention, there can be provided a honeycomb ceramics filter in which a predetermined trapping efficiency is maintained and a pressure loss by accumulation of the ash content is prevented from increasing to be not less than a predetermined pressure loss even with use for a long period.

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Claims

- 15 1. A honeycomb ceramics filter, characterized in that an average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

$$10 \geq W/X.$$

- 20 2. The honeycomb ceramics filter according to claim 1, wherein the average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

$$7 \geq W/X \geq 3.$$

- 25 3. The honeycomb ceramics filter according to claim 1, wherein the average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

$$5 \geq W/X \geq 3.$$

- 30 4. The honeycomb ceramics filter according to any one of claims 1 to 3, wherein a porosity is 55 to 75%.
- 35 5. The honeycomb ceramics filter according to any one of claims 1 to 4, wherein a ceramic contains cordierite and/or silicon carbide as a major component.
- 40 6. The honeycomb ceramics filter according to any one of claims 1 to 5, wherein a thermal expansion coefficient at 40 to 800°C is $1.0 \times 10^{-6}/^\circ\text{C}$ or less.
- 45 7. The honeycomb ceramics filter according to any one of claims 1 to 6, wherein the partition wall thickness W is 350 μm or less.
8. The honeycomb ceramics filter according to any one of claims 1 to 7, wherein a cell density is 250 cells/in² or more.

45 Amended claims under Art. 19.1 PCT

- 50 1. (Amended) A honeycomb ceramics filter, characterized in that the honeycomb ceramics filter contains cordierite as a major component, a thermal expansion coefficient at 40 to 800°C is $0.8 \times 10^{-6}/^\circ\text{C}$ or less, and an average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

$$10 \geq W/X.$$

- 55 2. The honeycomb ceramics filter according to claim 1, wherein the average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

7 ≥ W/X ≥ 3.

- 5 3. The honeycomb ceramics filter according to claim 1, wherein the average pore diameter X (μm) and partition wall thickness W (μm) of the filter satisfy the following relation:

5 ≥ W/X ≥ 3.

- 10 4. The honeycomb ceramics filter according to any one of claims 1 to 3, wherein a porosity is 55 to 75%.

5. (Deleted)

6. (Deleted)

- 15 7. (Amended) The honeycomb ceramics filter according to any one of claims 1 to 3, wherein the partition wall thickness W is 350 μm or less.

- 20 8. (Amended) The honeycomb ceramics filter according to any one of claims 1 to 3 and 7, wherein a cell density is 250 cells/in² or more.

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP02/02972

A. CLASSIFICATION OF SUBJECT MATTER
Int.Cl' B01D39/20

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
Int.Cl' B01D39/20Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
Jitsuyo Shinan Koho 1926-1996 Toroku Jitsuyo Shinan Koho 1994-2002
Kokai Jitsuyo Shinan Koho 1971-2002 Jitsuyo Shinan Toroku Koho 1996-2002Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
WPI (DIALOG)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 6-241018 A (Ibiden Co., Ltd.), 30 August, 1994 (30.08.94), Claims; Par. Nos. [0015] to [0028] (Family: none)	1-8
X	US 5846276 A (Matsushita Electric Industrial Co., Ltd.), 08 December, 1998 (08.12.98), Full text & JP 8-215522 A	1-4, 6-8
X	JP 9-71466 A (Denki Kagaku Kogyo Kabushiki Kaisha), 18 March, 1997 (18.03.97), Claims; examples (Family: none)	1, 2, 4, 5, 7

 Further documents are listed in the continuation of Box C. See patent family annex.

* Special categories of cited documents:	
"A" document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
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Date of the actual completion of the international search 17 June, 2002 (17.06.02)	Date of mailing of the international search report 02 July, 2002 (02.07.02)
Name and mailing address of the ISA/ Japanese Patent Office	Authorized officer

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Form PCT/ISA/210 (second sheet) (July 1998)

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP02/02972

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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X	JP 6-327921 A (Sumitomo Electric Industries, Ltd.), 29 November, 1994 (29.11.94), Claims; examples (Family: none)	1-3,4,7
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